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Neutron reflectometry by refractive encoding

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Abstract. There is a great deal of interest in expanding the technique of neutron reflectivity beyond static structural measurements of layered structures to kinetic studies. The time resolution for kinetic studies is limited only by the available neutron flux. On a continuous source of neutrons, such as a reactor, many reflectometers use the time-of-flight technique involving low-transmission choppers. This paper describes a real measurement involving prism refraction to deduce the wavelength in place of choppers thus providing a large gain in useful neutron flux and hence open up the possibility of sub-second kinetics studies.

Introduction

Specular neutron reflectometry is a technique used to establish the structure of layered materials on length scales varying from a fraction to hundreds of nm. Reflectivity is typically measured as a function of the momentum transfer, q of the neutron perpendicular to the surface. To vary q, either the reflection angle is varied at a fixed neutron wavelength or a range of wavelengths is used at a fixed angle. The second method is known as time of flight (TOF) as it involves periodically chopping the beam to measure the speed and hence wavelength. Figure 1 shows the best performance possible at the TOF instrument D17 [1] at the Institut Laue Langevin. The measurement time of 1 second was only possible by loosening the resolution to 4–10% depending on the wavelength. The advantage of using a prism would be to measure even faster but without the cost in resolution. The technique would be of equal value for experiments with sample areas much smaller than can be practically measured at present.

Previously, the method of using a prism to measure the wavelength has been described based on the measurements of the direct beam deflection [2]. Other white beam methods include oscillating the sample angle [3], elliptical mirrors [4] and field gradients [5]. Although dispersion by use of a magnetic-field gradient has the advantage of no losses due to absorption, the present practically achievable gradients cannot resolve wavelengths better than a material prism, particularly at short wavelengths. Here we describe the first experiment with a real sample demonstrating the feasibility of the technique.

The prism must be placed just after the sample and is calibrated by passing the direct beam through the prism alone to measure the deflection of the beam due to refraction. This can be done with the use of the choppers but, as the refractive dispersion is function of only the prism angle and the scattering length density, Nb of the prism material, both easily measurable quantities, it is not strictly necessary. The deflected angle, φ when striking the surface from the material side, is from Snells law:

$$\varphi = \cos^{-1}(n\cos(\alpha)) \quad \text{with} \quad n = \left(1 - \lambda^2 Nb/\pi\right)^{1/2} \tag{1}$$

where α is the angle of the beam to the prism surface n is the refractive index with λ the neutron wavelength.

With no choppers the intensity as a function of deflection is measured for the direct beam only. Then, with a sample in the direct beam, the prism plus detector needs to be rotated such that the sample reflection strikes the centre of the prism and this intensity is measured as a function of deflection, too. Figure 2 shows a sketch of the arrangement with the two intensities measured. The reflectivity is

$$R(\varphi) = \frac{I(\varphi)}{S(\varphi)}, \tag{2}$$

which is converted to $R(\lambda)$ via eq. (1) and then by applying Braggs law to $R(q)$.

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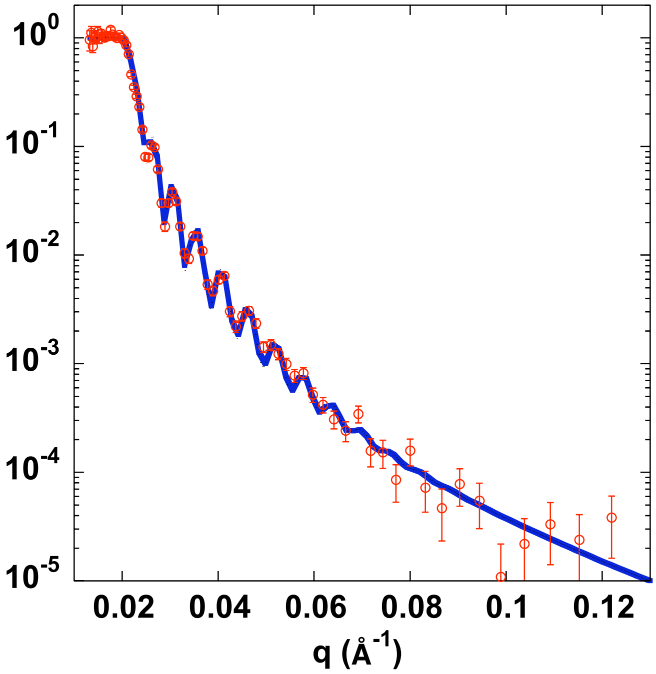

Fig. 1. One-second data from a $8 \times 5\,\mathrm{cm}^2$ sample of a 980 Å Ni layer on glass measured at D17 using TOF. The mean q resolution was 6%. The blue line is a fit to the data.

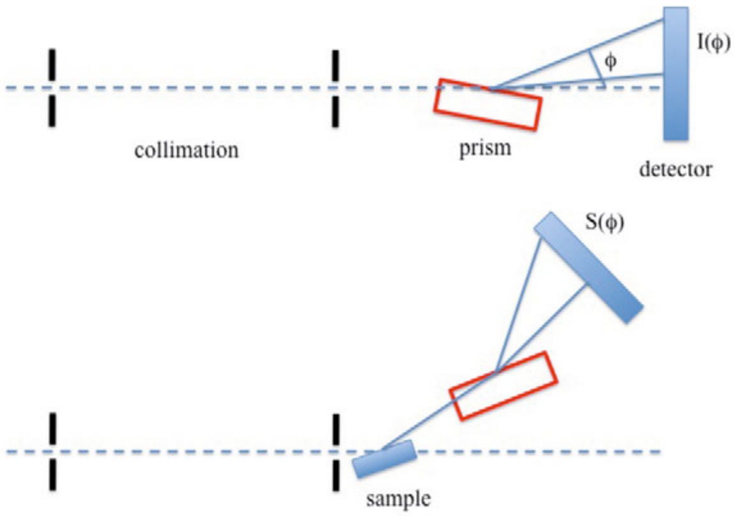

Fig. 2. A sketch of the experimental arrangement showing, at the top, the measurement of the direct beam intensity $I(\varphi)$. To measure the reflection from the sample $S(\varphi)$ both the prism and the detector are rotated about the sample as shown below.

Experimental results

The experiment was carried out on the AMOR [6] reflectometer at the Paul Scherrer Institute, Switzerland (PSI), which provides great flexibility due to its modular nature. The neutron beam was collimated by two 0.5 mm slits 1.583 m apart. The sample position was 0.31 m from the second slit and the prism was placed 0.742 m after the sample. Two different detectors were used, a conventional ^{3}He detector, 5.865 m from the sample, which could be used in conjunction with TOF with a spatial resolution of 2.4 mm FWHM and a CCD camera utilizing a neutron sensitive scintillater plate, 5.1 m from the sample with 0.2 mm resolution.

The sample was Ni/glass very similar but not identical to that used for the data in fig. 1. When measuring either the direct beam with no sample or the reflection from the sample the prism was displaced to keep the beam on the prism surface and the angle of the beam to the surface was kept constant at 0.14 deg. The prism was a simple rectangular block of MgF_2 ($50 \times 5 \times 140\,\mathrm{mm}^3$). The surface was polished with to a roughness of 10 Å and a flatness of

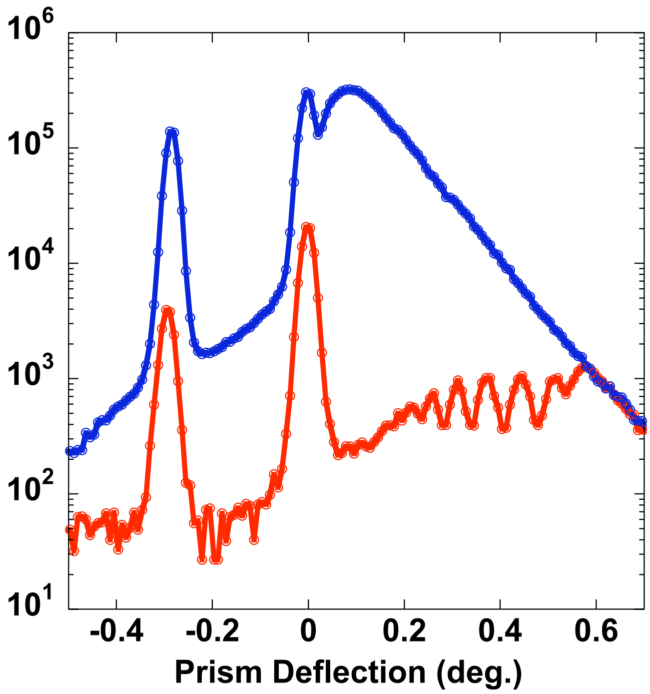

Fig. 3. Neutron counts from the main beam (blue) and reflection (red) after passing through the prism and counted on the ^{3}He detector. The peak on the left is the reflection from the prism surface and the central peak is the beam that missed the prism. To the right of this peak the different wavelengths are dispersed at different angles on the detector.

better than 0.005 deg. Counting times were 30 min for TOF, 3 min for the ^{3}He measurement and 1 h with the CCD. A longer counting time with the CCD was required as it had a very low detection efficiency $\sim 10\%$.

Figure 3 shows the signal on the detector as a function of position for both beams. The peak on the left is the reflection from the prism surface and the central peak is from neutrons that missed the prism surface. The right-hand side shows the dispersion of the beam thus resolving wavelength by deflection. The region of total reflection on the far right is clearly seen along with the Kiessig oscillations characteristic of the 1000 Å Ni layer on the sample. A background was removed from each data set, taken from the extreme left of the detector where the signals were nearly flat. In addition, the reflection data was multiplied by a factor to ensure the two signals were the same in the region of total reflection. Dividing one by the other and calculating the momentum transfer for each point knowing the sample reflection angle of 1 deg results in the desired reflectivity which is shown in fig. 4.

In order to check the previously published theory [2] the reflectivity was calculated using the Parratt formulism [7] from the known parameters of the sample, *i.e.* 1000 Å of Ni with 10 Å roughness on glass.

The FWHM resolution, Δq, was calculated for each point and then a Gaussian weighted moving average of this width was made at each point in q of the calculated reflectivity and then compared to the data. It was found that the assumption of linear dispersion with wavelength was not valid at low wavelengths so the width in q at each point was calculated as follows:

$$(\Delta q/q)^2 = (\Delta\lambda/\lambda)^2 + (\Delta\theta/\theta)^2\,, \tag{3}$$

where the two terms are the fractional spread in wavelength and angular variation of the incoming beam over the sample angle. From (1) the resolving power of the prism is

$$\mathrm{d}\varphi/\mathrm{d}\lambda = (\lambda N b\cos(\alpha))\,/\left(\pi\left(1-(n\cos(\alpha))^2\right)^{1/2}\right), \tag{4}$$

then the spread in wavelength $\Delta\lambda$ is

$$\Delta\lambda = (\mathrm{d}\varphi/\mathrm{d}\lambda)^{-1}\left[((s_1+s_2)\,/\,(2D_1))^2 + \Delta f^2 + (((L+\alpha)\,\varphi+\Delta x)\,/\,(2D_2))^2\right]^{1/2}, \tag{5}$$

where s_1 and s_2 are the first and second collimation slits separated by D_2, Δf is the angular variation of the prism surface, L is the illuminated length of the prism surface, Δx is the spatial resolution of the detector situated D_2 away from the prism. Measurements of the same sample by conventional TOF and with the prism using the two types of detector are shown in fig. 4 along with the same model smeared by the resolution as calculated above for the two

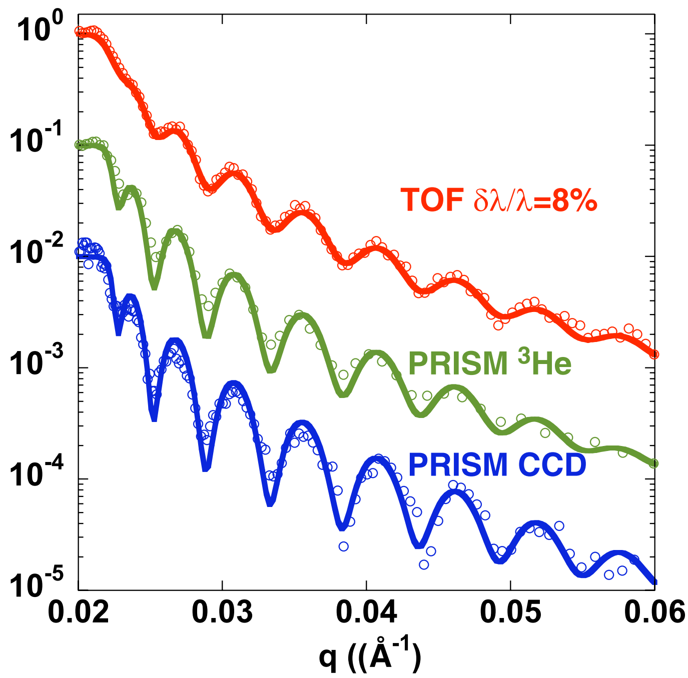

Fig. 4. Reflectivity as a function of momentum transfer for three different methods. The data in red are measured with TOF with 8% constant $\delta\lambda/\lambda$ resolution with the line a model of 1000 Å Ni on glass convolved by the appropriate resolution for each point using eq. (3). For the other two curves the resolution was calculated using eqs. (3) and (5). The curves have been displaced by factors of ten for visability.

Table 1. The fractional wavelength resolution and gain of the prism method over TOF for each instrument. The gains refer to the setup immediately above.

Method	$\Delta\lambda/\lambda$ (3.7 Å)	$\Delta\lambda/\lambda$ (11.0 Å)	Gain (3.7 Å)	Gain (11.0 Å)
TOF AMOR $\Delta x = 2.4\,\text{mm}$; $D_1 = 1.58\,\text{m}$; $D_2 = 5.1\,\text{m}$;	0.08	0.08	1	1
Prism $\Delta x = 2.4\,\text{mm}$; $D_1 = 1.58\,\text{m}$; $D_2 = 5.1\,\text{m}$;	0.087	0.032	33	3.2
Prism $\Delta x = 0.2\,\text{mm}$; $D_1 = 1.58\,\text{m}$; $D_2 = 5.0\,\text{m}$;	0.063	0.023	33	3.2
TOF D17 1s data $D_1 = 3.0\,\text{m}$; $D_2 = 3.0\,\text{m}$; $s_1 = 5$; $s_2 = 1.5$; $th = 1.5$ chopper opening 4 deg	0.10	0.04	1	1
Prism D17 $\Delta x = 0.2\,\text{mm}$; $D_1 = 3.0\,\text{m}$; $D_2 = 3.0\,\text{m}$; $s_1 = 2$; $s_2 = 0.5$; $th = 1.5$	0.11	0.035	8.0	2.0
TOF D17 $D_1 = 3.0\,\text{m}$; $D_2 = 3.0\,\text{m}$; $s_1 = 1$; $s_2 = 0.5$; $th = 1.0$ chopper opening 2.4 deg	0.065	0.031	1	1
Prism D17 $\Delta x = 0.2\,\text{mm}$; $D_1 = 3.0\,\text{m}$; $D_2 = 3.0\,\text{m}$; $s_1 = 1$; $s_2 = 0.5$; $th = 1.0$	0.065	0.028	92	22

different detector resolutions. It is clear that the model with resolution calculated using eqs. (5) and (3) matches well the data with the prism. The data shown corresponds to a wavelength range of 3.7–11.0 Å and the calculated fractional wavelength resolution for each case is shown in table 1.

The gain expressed is the gain in useful intensity of the prism method over the TOF methods at the same instrument. This is the ratio of the prism to chopper transmission if the slits are the same. The chopper resolution and transmission are expressed as follows:

$$\Delta\lambda/\lambda = d/D + (\psi/360)\,(\lambda_{\max}/\lambda) \qquad T = (d/D)\,(\lambda/\lambda_{\max}) + \psi/360, \tag{6}$$

Where d is the inter-chopper distance, D is the total TOF distance, ψ is the projected chopper opening and $\lambda_{\max}$ is the maximum wavelength, the TOF of which determines the chopper period. The transmission of the prism is a combination of three factors, losses due to absorption, incoherent scattering and reflection. These have been calculated for the known neutronic properties of MgF_2 to be 0.73 at 3.7 Å and 0.22 at 11.0 Å. The gain for the AMOR measurements is reflected in the factor of ten shorter counting time of the prism method over TOF on AMOR when using the ^{3}He detector. On D17, the projected gains are less compared to the 1 s data as, for approximately the same resolution, the TOF method on D17 can have wider slits so the gain is not just the ratio of the prism and chopper transmissions as the incoming intensity is not the same. However, if higher resolution than that used with the 1 s data is required, then the gains rise considerably. For example, we could get a much better resolution from the prism on D17 with $s_1 = 1.0$ and $s_2 = 0.5$ mm with $\Delta x = 0.2$ mm giving $\Delta\lambda/\lambda = 0.065$ at 3.7 Å and $\Delta\lambda/\lambda = 0.031$ at 11.0 Å. In this case, for $\theta = 1$ deg the angular resolution is nearly matched with the wavelength resolution at the longest wavelength. With the same slits with conventional TOF the wavelength resolutions are almost exactly matched by having a chopper opening of 2.4 deg giving corresponding transmissions of 0.0079 at 3.7 Å and 0.01 at 11.0 Å. In this case, the gains with the prism method are 92 at 3.7 Å and 22 at 11.0 Å.

Conclusion

The prism technique can be regarded as optimal for mid-range resolution. Due to practical limits in detector resolution and collimation, a resolution of $\Delta\lambda/\lambda < 0.05$ at short wavelengths is probably not practical. However, in the range $0.05 < \Delta\lambda/\lambda < 0.15$, there are considerable gains in useful intensity, the high limit being determined by the ability of the conventional technique to loosen the angular resolution which cannot be done with the prism. For both AMOR and D17 the prism method could be used as an option with the principle investment being an area detector with $\Delta x = 0.2$ required in one dimension only (the other dimension can be summed). We envisage this technique to be an important tool in increasing-time resolution for kinetics studies and in measuring very small samples where intermediate resolution is still required.

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